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Structural and Chemical Controls of Radiolytic H_2

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Water radiolysis, driven by the decay of ^{238}U, ^{235}U, ^{232}Th & ^{40}K, is a ubiquitous source of H_2 in the Earth's crust. However, current production rate estimates rely on deterministic models that assume homogeneous, intergranular porosity—a geometry that fails to capture the complexity of fractured crystalline basement rocks [1-4]. Furthermore, while fluid composition is known to influence radiolytic yields (G-values), these effects remain poorly constrained for deep crustal fluids.

In this study, we couple Monte Carlo particle transport simulations with experimental irradiation data to refine crustal H_2 generation rates. We apply the Geant4 toolkit to model the spatial distribution of radiation energy deposition within fractured 3D rock volumes. This represents the first application of Geant4 to simulate energy partitioning between the mineral matrix and the aqueous phase within fracture geometries while simultaneously tracking the production of radiogenic and nucleogenic isotopes.

To validate the simulation physics and resolve chemical controls, we performed α and γ irradiations of both synthetic and natural basement brines containing variable concentrations of dissolved scavengers. H_2 generation was quantified via micro-gas chromatography to determine $G(H_2)$ values specific to brine composition.

Preliminary results show that H_2/He ratios from average continental crust can vary significantly (2.3x) as a function of volumetric fracture intensity (P32), independent of porosity. This was attributed to enhanced energy deposition efficiency within the fluid filled fractures. Furthermore, $G(H_2)$ was found to be sensitive to fluid composition with certain hypersaline brines exhibiting a 1.8-fold increase in H_2 yield relative to seawater. Validation of the particle transport model against Fricke dosimetry showed a mean error of <5%. Integrating the experimentally derived H_2 yields with numerically modelled energy deposition rates provides a validated approach for quantifying H_2 generation rates and interpreting H_2/He ratios in the continental crust.

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